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Batch Experiments for Desorption of Plutonium and Americium in Contaminated Soil from the Rocky Flats Plant

Ines R. Triay and Gary W. Loge

Report for Rocky Flats

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Introduction

The soil at a former waste storage area at the Rocky Flats Plant (RFP) of DOE, which was originally used to store waste in metal drums, is contaminated with plutonium and americium. Contamination has been caused by drums that corroded and leaked waste into the soil. Since the discovery of this leakage, the drums have been removed and the soil in the area has been covered by asphalt.

A series of experiments were performed at Los Alamos National Laboratory (LANL) to identify leaching schemes that would be suitable for leaching plutonium and americium from the contaminated soil. The procedure for these batch desorption experiments on RFP soil (contaminated with plutonium and americium) is described and preliminary analysis of the results is presented.

Actinides (such as Pu, Am, and U) tend to have a strong affinity for the minerals in most soils (Thomas, 1987 and Triay et al., 1991). The mechanisms that normally dominate radionuclide sorption are surface complexation (Combes et al , 1992) and ion exchange (Triay and Rundberg, 1989 and 1987) Since the radioactive metals (that contaminated the RFP soil) are Lewis acids (i.e., acquire electrons to reach an inert state), complexants that act as Lewis bases (i.e , have electron pairs that can be shared with the metal) can be utilized to leach Pu and Am from contaminated soils.

The effectiveness of complexants to remove plutonium and americium from soils depends on the chemical form of these metals in the contaminated area Brainard et al. (1992) have shown that siderophores can effectively dissolve plutonium oxide. Pimpl and Schucttelkopf (1991) report that Pu, Am, and Cm in soil columns, contaminated with 5 mCi of each actinide near the surface, were mobilized and migrated with an irrigation solution containing 0 1M DTPA. After elution of the irrigation solution less than 3 pCi/g of activity was found in the soil. Lee and Marsh (1992) have reported that a significant amount of uranium can be extracted from Fernald soils utilizing citric acid and carbonate.

The choice of leaching agents (used in this study) was made taking into consideration previous results obtained with plutonium-contaminated soil from the Nevada Test Site (NTS) The NTS experiments involved air-drying

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soil from the NTS, sieving the soil to recover different size fractions, and alpha counting the soil to identify the most contaminated size fraction The results of these experiments indicated that most of the plutonium was associated with soil particles less than 53 μm in diameter Consequently, all the leaching experiments were performed with soil from the NTS with a particle size of less than 53 μm.

Leaching experiments involved adding a solution containing a complexing agent to the contaminated soil, mixing the two phases, separating the phases, and determining the amount of plutonium in each phase. The experimental parameters of the NTS leaching experiments were as follows The soil to solution ratio was 1:8, the contact time between the solution and solid phase was 24 hours, the concentration of the complexants in all solutions was 0 1 M, and the separation of phases was conducted by centrifugation The initial ^{239}Pu concentration of the NTS soil was 1,400 pCi/g All the NTS experiments were conducted at 20°C The results of these experiments are given in Table 1.

Table 1. Results of Plutonium-Leaching Experiments from the NTS Soils

Extractant	% Removal	Starting pH	Equilibrium pH
EDTA / 3% H_2O_2	22	4.4	7 7
EDTA	21	4 5	5 6
EDTA / 1 eq NaOH	14	8 0	8 6
EDTA / 2 eq NaOH	9	11 0	10.9
EDTA / 3 eq NaOH	5	12.3	12.0
Citric Acid / 3% H_2O_2	40	1.9	3.7
Citric Acid / 1 eq HNO_3	18	1 4	3 1
Citric Acid	16	2 2	3 8
Citric Acid / 1 eq NaOH	12	3 8	5 2
Citric Acid / 2 eq NaOH	10	5 1	8 2
Citric Acid / 3 eq NaOH	2	11.6	10 2
Citric Acid / 4 eq NaOH	0	12.3	12 3
Na_2EGTA	13	7.4	6 3
Na_2EGTA / 2 eq NaOH	45	12.0	11 5

The results shown in Table 1 indicate that simple complexants may be able to desorb plutonium from plutonium-contaminated soils and that adding an oxidizing or reducing agent may assist in the plutonium-leaching process.

Experimental Procedure

Several sealed containers of soil, taken from the contaminated RFP site, were sent to LANL for the soil extraction experiments The results described were obtained using samples from Container No DRII 86700 The soil from this container was air dried, crushed, and then sieved into a series of size ranges using progressively finer mesh sieves Soil with a particle size less than 53 µm in diameter was used for the RFP desorption experiments

This choice was based on the amount of alpha activity associated with each size fraction of the RFP soil (determined with a gas proportional alpha counter). As shown in Table 2, the largest amount of alpha activity is associated with the particles smaller than 53 µm in diameter

Table 2. Alpha Activity Associated with Size Fractions of the RFP Soil

Particle Size (µm)	CPM/g
<53	2,300
53 - 75	1,200
75 - 250	890
250 - 500	300
500 - 2,000	140
> 2,000	140

All desorption experiments were performed in duplicate. The desorption experiment consists of: 1) weighing 2 5 g of the RFP sieved soil (with a particle size of <53 µm) into an Oak Ridge centrifuge tube, 2) adding 20 ml of a freshly prepared extractant solution to the soil in the tube and capping the tube tightly, 3) placing the tube in an orbital shaker for 24 hours, 4) centrifuging the sample for one hour at 12,000 rpm (28,000 g), 5) decanting the liquid from the solid into a fresh centrifuge tube, 6) centrifuging the decanted liquid for one hour at 12,000 rpm, 7) pipetting 10 ml of the centrifuged liquid into a third centrifuge tube for a final two hour centrifugation cycle at 12,000 rpm, 8) analyzing 5 ml of the centrifuged liquid for ^{239}Pu and ^{241}Am content (using alpha and gamma spectrometry, respectively), 9) weighing the wet solid sample (left from step 5), 10) drying the wet solid under a heat lamp for 48 hours, then weighing again to obtain the weight of the residual extractant liquid from the decanting procedure, and 11) analyzing the dried soil samples for ^{239}Pu and ^{241}Am content using gamma spectrometry.

Gamma spectrometry analysis was performed using a 180-cc HPGe detector with a well (1 cm in diameter and 5 cm deep) into which a Plexiglas capsule with the sample is placed. This geometry permits capturing about 90% of

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^{241}Am gammas and about 60% of the ^{239}Pu gammas All dry solid samples had a very intense ^{241}Am 59 5 KeV peak and a much weaker, but unmistakably identifiable ^{239}Pu 129 5 KeV peak The reason why the Pu peak is weaker than the Am one is that only 0 006 alpha-decays of ^{239}Pu are accompanied by a 129 5 KeV photon whereas about half of ^{241}Am decays give a 59 5 KeV photon Each dry solid sample was measured for about 4 hours The ^{239}Pu concentration in the solid samples was measured by gamma spectrometry with 20-30% accuracy in most cases; the ^{241}Am concentration was measured with much better accuracy in both solid and liquid samples

The extractant solutions were prepared as follows.

0 100 M Citric Acid: 5 25 g of Citric acid monohydrate (FW = 210 0) were dissolved in enough distilled water to produce 250 mL of solution

0.100 M Citric Acid / 2 eq NaOH· 5 25 g of Citric acid monohydrate (FW = 210.0) and 50.0 mL of 1 00 M NaOH were dissolved in enough distilled water to produce 250. mL of solution.

0 100 M Citric Acid in 3% H_2O_2: 2.101 g of citric acid monohydrate (FW = 210.1) were dissolved in enough 3% H_2O_2 to produce 100 mL of solution

0.100 M Citric Acid / 0.01 M Fe^{3+} in 3% H_2O_2. 2.1 g of Citric acid monohydrate (FW = 210 0) and 0 270 g of ferric chloride hexahydrate (FW = 270.30) were dissolved in enough 3% hydrogen peroxide to produce 100. mL of solution.

0.100 M Citric Acid in 5 % NaOCl: 2.101 g of citric acid monohydrate (FW = 210 1) were dissolved in enough commercial bleach to produce 100. mL of solution.

0.100 M Citric Acid / 0.100 M $Na_2S_2O_8$: 2 101 g of citric acid monohydrate (FW = 210.1) and 2.380 g of sodium persulfate (FW = 238.0) were dissolved in enough distilled water to produce 100. mL of solution.

0.100 M Citric Acid / 0.100 M $Na_2S_2O_4$. 2.101 g of citric acid monohydrate (FW = 210.1) and 1.741 g of sodium dithionite (FW = 174.1) were dissolved in enough distilled water to produce 100. mL of solution.

0.100 M Citric Acid / 0.100 M $NH_2OH \cdot HCl$: 2.101 g of citric acid monohydrate (FW = 210.1) and 0 695 g of hydroxylamine hydrochloride (FW = 69.49) were dissolved in enough distilled water to produce 100. mL of solution.

0.100 M Citric Acid / 0.100 M Ascorbic Acid: 2.101 g of citric acid monohydrate (FW = 210.1) and 1 761 g of ascorbic acid (FW = 176.1) were dissolved in enough distilled water to produce 100. mL of solution.

0 100 M Sodium Citrate in 3% H_2O_2 2 941 g of sodium citrate dihydrate (FW = 294.1) were dissolved in enough 3% H_2O_2 to produce 100 mL of solution

0 100 M Sodium Citrate / 0 01 M Fe^{3+} in 3% H_2O_2 2 941 g of sodium citrate dihydrate (FW = 294 1) and 0 270 g of ferric chloride hexahydrate (FW = 270 30) were dissolved in enough 3% H_2O_2 to produce 100 mL of solution

0.100 M Sodium Citrate in 5% NaOCl 2 941 g of sodium citrate dihydrate (FW = 294 1) were dissolved in enough commercial bleach to produce 100 mL of solution

0.100 M Sodium Citrate / 0.100 M $Na_2S_2O_8$ 2 941 g of sodium citrate dihydrate (FW = 294.1) and 2 380 g of sodium persulfate (FW = 238 0) were dissolved in enough distilled water to produce 100 mL of solution.

0.100 M Sodium Citrate / 0.100 M $Na_2S_2O_4$ 2 941 g of sodium citrate dihydrate (FW = 294.1) and 1 741 g of sodium dithionite (FW = 174 1) were dissolved in enough distilled water to produce 100 mL of solution

0 100 M Sodium Citrate / 0.100 M NH_2OH HCl· 2 941 g of sodium citrate dihydrate (FW = 294.1) and 0 695 g of hydroxylamine hydrochloride (FW = 69.49) were dissolved in enough distilled water to produce 100 mL of solution.

0.100 M Sodium Citrate / 0.100 M Ascorbic Acid 2 941 g of sodium citrate dihydrate (FW = 294.1) and 1.761 g of ascorbic acid (FW = 176 1) were dissolved in enough distilled water to produce 100 mL of solution

0.100 M Na_2EDTA: 9 31 g of the Disodium salt of ethylenediamine tetraacetic acid dihydrate (FW = 372.2) were dissolved in enough distilled water to produce 250. mL of solution.

0.100 M Na_2EDTA / 1 eq NaOH: 9 31 g of the Disodium salt of ethylenediamine tetraacetic acid dihydrate (FW = 372.2) and 25.0 mL of 1.00 M NaOH were dissolved in enough distilled water to produce 250 mL of solution.

0.100 M Na_2EDTA in 3% H_2O_2: 3 722 g of disodium EDTA (FW = 372 24) were dissolved in enough 3% hydrogen peroxide to produce 100. mL of solution.

0.100 M Na_2EDTA / 0.01 M Fe^{3+} in 3% H_2O_2· 3 722 g of disodium EDTA (FW = 372.24) and 0.270 g of ferric chloride hexahydrate (FW = 270 30) were dissolved in enough 3% hydrogen peroxide to produce 100. mL of solution

0.100 M Na_2EGTA: 3 80 g of Ethyleneglycol-O,O'-bis(2-aminoethyl)-N,N,N',N'-tetraacetic acid (FW = 380 4) and 20 0 mL of 1 00 M NaOH were dissolved in enough distilled water to produce 100 mL of solution.

0 100 M DTPA / 3 eq NaOH 9 83 g of Diethylenetriaminepentaacetic acid (FW = 393 3) and 75 0 mL of 1 00 M NaOH were dissolved in enough distilled water to produce 250 mL of solution

0 100 M $(NH_4)_2CO_3$. 0 781 g of ammonium carbamate (FW = 78 07) were dissolved in enough distilled water to produce 100 mL of solution Ammonium carbamate hydrolyzes instantly to ammonium carbonate (FW = 96 0) in water Ammonium carbamate should be exposed to the atmosphere the *least* amount possible

0 100 M Na_2CO_3 in 3% H_2O_2: 1 060 g of sodium carbonate (FW = 106 0) were dissolved in enough 3% H_2O_2 to produce 100. mL of solution

0.100 M Na_2CO_3 / 0.01 M Fe^{3+} in 3% H_2O_2· 1 060 g of sodium carbonate (FW = 106.0) and 0.199 g of ferrous chloride tetrahydrate (FW = 198 8) were dissolved in enough 3% hydrogen peroxide to produce 100 mL of solution

0.100 M Na_2CO_3 in 5 % NaOCl: 1 060 g of sodium carbonate (FW = 106 0) were dissolved in enough commercial bleach to produce 100 mL of solution

0 100 M Na_2CO_3 / 0.100 M $Na_2S_2O_8$: 1.060 g of sodium carbonate (FW = 106 0) and 2 380 g of sodium persulfate (FW = 238.0) were dissolved in enough distilled water to produce 100. mL of solution.

0.100 M Na_2CO_3 / 0.100 M $Na_2S_2O_4$: 1 060 g of sodium carbonate (FW = 106 0) and 1 741 g of sodium dithionite (FW = 174 1) were dissolved in enough distilled water to produce 100. mL of solution.

0 100 M Na_2CO_3 / 0.100 M $NH_2OH \cdot HCl$: 1 060 g of sodium carbonate (FW = 106 0) and 0 695 g of hydroxylamine hydrochloride (FW = 69 49) were dissolved in enough distilled water to produce 100. mL of solution.

0.100 M Na_2CO_3 / 0.100 M Ascorbic Acid: 1 060 g of sodium carbonate (FW = 106 0) and 1.761 g of ascorbic acid (FW = 176.1) were dissolved in enough distilled water to produce 100. mL of solution.

0.100 M $NaHCO_3$: 0 840 g of sodium bicarbonate (FW = 84 01) were dissolved in enough distilled water to produce 100. mL of solution.

0.100 M $NaHCO_3$ in 3% H_2O_2: 0 840 g of sodium bicarbonate (FW = 84 01) were dissolved in enough 3% H_2O_2 to produce 100. mL of solution.

0.100 M $NaHCO_3$ / 0.01 M Fe^{3+} in 3% H_2O_2: 0.840 g of sodium bicarbonate (FW = 84.01) and 0.199 g of ferrous chloride tetrahydrate (FW = 198.8) were dissolved in enough 3% hydrogen peroxide to produce 100 mL of solution.

0.100 M $NaHCO_3$ in 5% NaOCl: 0.840 g of sodium bicarbonate (FW = 84.01) were dissolved in enough commercial bleach to produce 100. mL of solution

0 100 M $NaHCO_3$ / 0 100 M $Na_2S_2O_8$ 0 840 g of sodium bicarbonate (FW = 84 01) and 2 38 g of sodium persulfate (FW = 238 0) were dissolved in enough distilled water to produce 100 mL of solution

0 100 M $NaHCO_3$ / 0.100 M $Na_2S_2O_4$ 0 840 g of sodium bicarbonate (FW = 84 01) and 1 74 g of sodium dithionite (FW = 174 1) were dissolved in enough distilled water to produce 100. mL of solution.

0 100 M $NaHCO_3$ / 0 100 M NH_2OH HCl 0 840 g of sodium bicarbonate (FW = 84 01) and 0.695 g of hydroxylamine hydrochloride (FW = 69 49) were dissolved in enough distilled water to produce 100 mL of solution

0.100 M $NaHCO_3$ / 0 100 M Ascorbic Acid· 0 840 g of sodium bicarbonate (FW = 84 01) and 1 761 g of ascorbic acid (FW = 176 1) were dissolved in enough distilled water to produce 100 mL of solution.

0.100 M Nitrilotriacetic Acid / 2 eq NaOH· 1 911 g of nitrilotriacetic acid (FW = 191 1) and 20 0 mL of 1 00 M NaOH were dissolved in enough distilled water to produce 100. mL of solution.

0.100 M Nitrilotriacetic Acid / 3 eq NaOH: 1 911 g of nitrilotriacetic acid (FW = 191.1) and 30.0 mL of 1.00 M NaOH were dissolved in enough distilled water to produce 100. mL of solution.

0.100 M TETA in 3% H_2O_2: 1.462 g of triethylenetetramine (FW = 146.2) were dissolved in enough 3% hydrogen peroxide to produce 100. mL of solution

0.100 M TETA / 0 01 M Fe^{3+} in 3% H_2O_2· 1 462 g of triethylenetetramine (FW = 146.2) and 0.270 g of ferric chloride hexahydrate (FW = 270 30) were dissolved in enough 3% hydrogen peroxide to produce 100. mL of solution

0.100 M TETA / 0.100 M $NaHCO_3$ in 3% H_2O_2 1 462 g of triethylenetetramine (FW = 146.2) and 0.840 g of sodium bicarbonate (FW = 84.01) were dissolved in enough 3% hydrogen peroxide to produce 100. mL of solution.

0.100 M TETA / 0.100 M $NaHCO_3$ in 5% NaOCl: 1 462 g of triethylenetetramine (FW = 146 2) and 0 840 g of sodium bicarbonate (FW = 84.01) were dissolved in enough commercial bleach to produce 100 mL of solution.

0.100 M TETA / 0.100 M $NaHCO_3$ / 0.100 M $Na_2S_2O_8$: 1 462 g of triethylenetetramine (FW = 146.2), 0 840 g of sodium bicarbonate (FW = 84 01), and 2.380 g of sodium persulfate (FW = 238.0) were dissolved in enough distilled water to produce 100. mL of solution.

0.100 M TETA / 0.100 M $NaHCO_3$ / 0 100 M $Na_2S_2O_4$: 1.462 g of triethylenetetramine (FW = 146 2), 0 840 g of sodium bicarbonate (FW = 84.01), and 1.741 g sodium dithionite (FW = 174 1) were dissolved in enough distilled water to produce 100. mL of solution

0.100 M TETA / 0 100 M $NaHCO_3$ / 0 100 M NH_2OH HCl 1 462 g of triethylenetetramine (FW = 146 2), 0 840 g of sodium bicarbonate (FW = 84 01), and 0 695 g of hydroxylamine hydrochloride (FW = 69 49) were dissolved in enough distilled water to produce 100 mL of solution

0 100 M TETA / 0.100 M $NaHCO_3$ / 0 100 M Ascorbic Acid. 1 462 g of triethylenetetramine (FW = 146 2), 0 840 g of sodium bicarbonate (FW = 84 01), and 1 761 g of ascorbic acid (FW = 176.1) were dissolved in enough distilled water to produce 100. mL of solution.

0.100 M Tartaric Acid / 1 eq NaOH: 3 75 g of Tartaric acid (FW = 150 1) and 25 0 mL of 1.00 M NaOH were dissolved in enough distilled water to produce 250. mL of solution.

0.100 M Triethyleneglycol / 0.100 M $NaHCO_3$: 1 502 g of triethyleneglycol (FW = 150 2) and 0.840 g of sodium bicarbonate (FW = 84 01) were dissolved in enough distilled water to produce 100 mL of solution.

0 100 M Triethyleneglycol in 3% H_2O_2. 1 502 g of triethyleneglycol (FW = 150.2) were dissolved in enough 3% hydrogen peroxide to produce 100 mL of solution.

0.100 M Triethyleneglycol / 0.01 M Fe^{3+} in 3% H_2O_2: 1 502 g of triethylene glycol (FW = 150.2) and 0.270 g of ferric chloride hexahydrate (FW = 270 30) were dissolved in enough 3% hydrogen peroxide to produce 100 mL of solution.

0 100 M Triethyleneglycol, DME / 0.100 M $NaHCO_3$: 1.782 g of triethyleneglycol, dimethyl ether (FW = 178.2) and 0.840 g of sodium bicarbonate (FW = 84.01) were dissolved in enough distilled water to produce 100. mL of solution.

0.100 M Triethyleneglycol, DME in 3% H_2O_2: 1 782 g of triethyleneglycol, dimethyl ether (FW = 178.2) and 0.199 g of ferric chloride tetrahydrate (FW = 198.8) were dissolved in enough 3% hydrogen peroxide to produce 100. mL of solution.

0.100 M Triethyleneglycol, DME in 5% NaOCl: 1.782 g of triethyleneglycol, dimethyl ether (FW = 178.2) were dissolved in enough commercial bleach to produce 100. mL of solution.

0.100 M Triethyleneglycol, DME / 0.100 M $Na_2S_2O_8$: 1.782 g of triethyleneglycol, dimethyl ether (FW = 178 2) and 2.380 g of sodium persulfate (FW = 238.0) were dissolved in enough distilled water to produce 100. mL of solution.

0.100 M Triethyleneglycol, DME / 0.100 M $Na_2S_2O_4$: 1.782 g of triethyleneglycol, dimethyl ether (FW = 178.2) and 1.741 g sodium dithionite (FW = 174.1) were dissolved in enough distilled water to produce 100. mL of solution.

0 100 M Triethyleneglycol, DME / 0 100 M NH_2OH HCl 1 782 g of triethyleneglycol, dimethyl ether (FW = 178 2) and 0 695 g of hydroxylamine hydrochloride (FW = 69 49) were dissolved in enough distilled water to produce 100 mL of solution

0 100 M Triethyleneglycol, DME / 0 100 M Ascorbic Acid 1 782 g of triethyleneglycol, dimethyl ether (FW = 178 2) and 1 761 g of ascorbic acid (FW = 176 1) were dissolved in enough distilled water to produce 100 mL of solution

0.100 M Tiron. 7 86 g of Tiron (FW = 314 2) were dissolved in enough distilled water to produce 250 mL of solution

0.100 M HNO_3: 10 0 mL of stock 1 00 M nitric is diluted with enough distilled water to produce 100. mL of solution. [Preparation of stock 1 00 M HNO_3 62.9 mL of concentrated nitric acid is diluted with enough distilled water to produce 1 00 L of solution.]

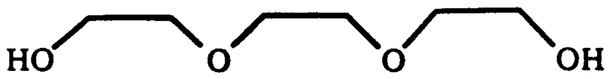
5% NaOCl: Commercial bleach

0.100 M $NH_2OH \cdot HCl$: 0 695 g of hydroxylamine hydrochloride (FW = 69 49) were dissolved in enough distilled water to produce 100 mL of solution

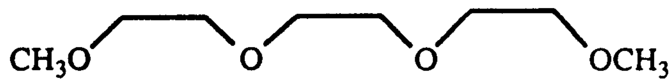
0.100 M Ascorbic Acid: 1 761 g ascorbic acid (FW = 176 1) were dissolved in enough distilled water to produce 100 mL of solution.

The chemical structures of the complexants and the oxidizing and the reducing agents (used in the leaching schemes) are as follows.

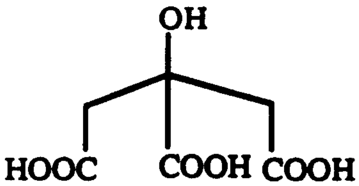
Rocky Flats Soil Extractants



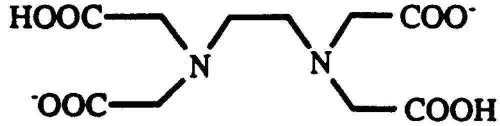
Triethyleneglycol
TEG



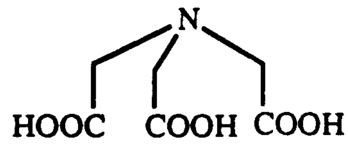
Triethyleneglycol dimethyl ether
TEG, DME



Citric Acid

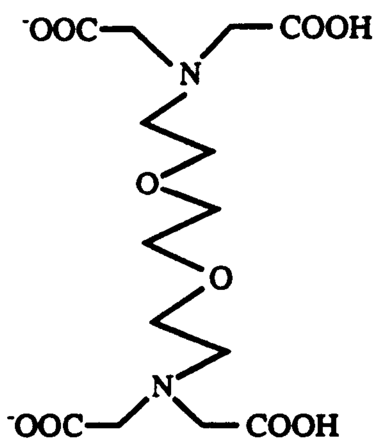


Na_2EDTA
(Disodium Salt)

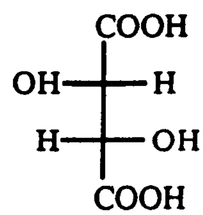


NTA
Nitrilotriacetic Acid

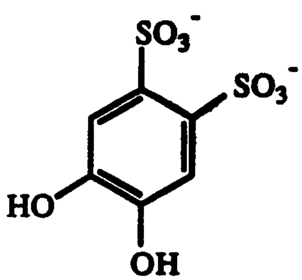
TETA
Triethylenetetramine



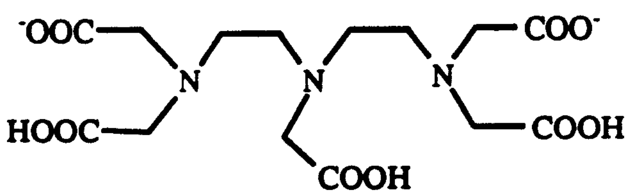
Na_2EGTA
Ethyleneglycol-O,O'-bis(aminoethyl)-N,N,N',N'-tetraacetic acid
(Disodium Salt)



Tartaric Acid



Tiron



Na_2DTPA
Diethylenetriaminepentaacetic acid
(Disodium Salt)

011

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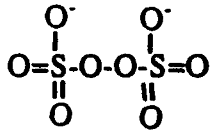
Rocky Flats Soil Redox Agents

Oxidizing Agents



persulfate (sodium salt)

H-O-O-H

hydrogen peroxide

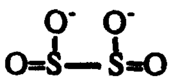
NaOCl

bleach (sodium hypochlorite)

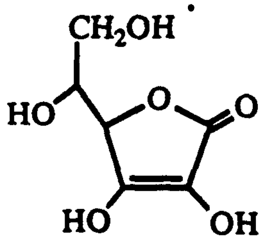
O_3

ozone

Reducing Agents



dithionite (sodium salt)



ascorbic acid

$NH_2OH \quad HCl$

hydroxylamine hydrochloride

H_2S

hydrogen sulfide

Results

The results of the leaching experiments described in the experimental section are presented using the gamma spectrometric analyses of the solid samples only. The assumptions made in reporting these results are: 1) the initial concentration of Am and Pu in the soil is homogeneous, and 2) the fraction of extracted ^{241}Am and ^{239}Pu is the same as the total fraction of Am and Pu extracted (i.e., all isotopes of Pu and Am have the same chemical behavior) The first assumption is probably not reasonable. A better estimate of the extraction efficiency will be presented in a future report by determining the amount of ^{241}Am and ^{239}Pu in the liquid samples using alpha and gamma spectrometry, respectively.

The results of analyzing four soil solid samples prior to treatment are given in Table 3. These solid samples were analyzed using gamma spectrometry (as described in the experimental section). The soil analyzed (and used for the treatment studies) had a particle size smaller than 53 μm. The results presented in Table 4 assumed that the initial concentration of ^{241}Am and ^{239}Pu in the soil treated was 2, 200 and 7,800 pCi/g, respectively.

Table 3: Initial Am and Pu Contamination in RFP Soil (with particle size < 53 μm in diameter)

	^{241}Am, pCi/g	Error in ^{241}Am Analysis, pCi/g	^{239}Pu, pCi/g	Error in ^{239}Pu Analysis, pCi/g
Trial #1	1,971	101	6,647	592
Trial #2	2,276	119	8,231	889
Trial #3	2,253	118	8,100	1,296
Trial #4	2,269	121	8,285	911
Average	2,192		7,816	

Table 4. Results of Pu and Am Leaching Experiments

Extractant	% Am extracted	% Pu extracted
0.1 M Citric Acid	13%	24%
0.1 M Citric Acid / 2 eq NaOH	39%	18%
0.1 M Citric Acid / 3% H_2O_2	14%	42%
0.1 M Citric Acid / 0.01 M Fe^{3+} / 3% H_2O_2	20%	39%
0.1 M Citric Acid / 5% NaOCl	3%	32%
0.1 M Citric Acid / 0.1 M $Na_2S_2O_8$	7%	30%
0.1 M Citric Acid / 0 1 M $Na_2S_2O_4$	15%	51%
0.1 M Citric Acid / 0.1 M $NH_2OH{\cdot}HCl$	2%	25%
0.1 M Citric Acid / 0.1 M Ascorbic Acid	16%	18%
0.1 M Sodium Citrate / 3% H_2O_2	42%	19%
0.1 M Sodium Citrate / 0.01 M Fe^{3+} / 3% H_2O_2	43%	43%
0.1 M Sodium Citrate / 5% NaOCl	12%	0%
0.1 M Sodium Citrate / 0.1 M $Na_2S_2O_8$	45%	29%
0.1 M Sodium Citrate / 0.1 M $Na_2S_2O_4$	57%	61%

0 1 M Sodium Citrate/ 0 1 M NH_2OH HCl	41%	36%
0 1 M Sodium Citrate / 0 1 M Ascorbic Acid	58%	51%
0 1 M EDTA / 2 eq NaOH	39%	8%
0.1 M EDTA / 3 eq NaOH	45%	8%
0.1 M EDTA / 3% H_2O_2	47%	26%
0 1 M EDTA / 0 01 M Fe^{3+} / 3% H_2O_2	0%	0%
0.1 M EGTA / 2 eq NaOH	30%	7%
0.1 M DTPA / 3 eq NaOH	46%	25%
0.1 M $(NH_4)_2CO_3$	0%	6%
0.1 M Na_2CO_3 / 3% H_2O_2	0%	0%
0.1 M Na_2CO_3 / 0.01 M Fe^{3+} / 3% H_2O_2	0%	14%
0.1 M Na_2CO_3 / 5% NaOCl	11%	28%
0.1 M Na_2CO_3 / 0.1 M $Na_2S_2O_8$	0%	10%
0.1 M Na_2CO_3 / 0.1 M $Na_2S_2O_4$	0%	3%
0 1 M Na_2CO_3 / 0 1 M $NH_2OH{\cdot}HCl$	0%	0%
0.1 M Na_2CO_3 / 0.1 M Ascorbic Acid	27%	20%
0.1 M $NaHCO_3$	2%	12%
0.1 M $NaHCO_3$ / 3% H_2O_2	0%	0%
0.1 M $NaHCO_3$ / 0.01 M Fe^{3+} / 3% H_2O_2	0%	5%
0.1 M $NaHCO_3$ / 5% NaOCl	13%	14%
0.1 M $NaHCO_3$ / 0.1 M $Na_2S_2O_8$	0%	10%
0.1 M $NaHCO_3$ / 0.1 M $Na_2S_2O_4$	0%	4%
0.1 M $NaHCO_3$ / 0.1 M $NH_2OH{\cdot}HCl$	0%	2%
0.1 M $NaHCO_3$ / 0.1 M Ascorbic Acid	22%	11%
0.1 M Nitrilotriacetic Acid / 2 eq NaOH	48%	2%

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0.1 M Nitrilotriacetic Acid / 3 eq NaOH	41%	23%
0.1 M TETA / 3% H_2O_2	0%	9%
0 1 M TETA / 0.1 M $NaHCO_3$ / 3% H_2O_2	0%	0%
0 1 M TETA / 0.1 M $NaHCO_3$ / 5% NaOCl	9%	13%
0 1 M TETA / 0.1 M $NaHCO_3$ / 0.1 M $Na_2S_2O_8$	5%	18%
0 1 M TETA / 0.1 M $NaHCO_3$ / 0.1 M $Na_2S_2O_4$	0%	0%
0.1 M TETA / 0.1 M $NaHCO_3$ / 0.1 M $NH_2OH{\cdot}HCl$	0%	12%
0.1 M TETA / 0.1 M $NaHCO_3$ / 0.1 M Ascorbic Acid	3%	11%
0.1 M Tartaric Acid / 1 eq NaOH	0%	5%
0.1 M TEG / 0.1 M $NaHCO_3$	0%	0%
0.1 M TEG / 3% H_2O_2	0%	0%
0.1 M TEG / 0.01 M Fe^{3+} / 3% H_2O_2	4%	3%
0.1 M TEG-DME / 0.1 M $NaHCO_3$	0%	0%
0.1 M TEG-DME / 5% NaOCl	0%	8%
0.1 M TEG-DME / 0.1 M $Na_2S_2O_8$	0%	0%
0.1 M TEG-DME / 0.1 M $Na_2S_2O_4$	0%	0%
0.1 M TEG-DME / 0.1 M $NH_2OH{\cdot}HCl$	0%	3%
0.1 M TEG-DME / 0.1 M Ascorbic Acid	0%	0%
0.1 M Tiron	0%	24%
0.1 M Nitric Acid	7%	0%
5% NaOCl	15%	8%
0.1 M $NH_2OH{\cdot}HCl$	0%	0%
0.1 M Ascorbic Acid	0%	0%

Conclusions

1) The maximum amount of contamination in the RFP soils is associated with the smallest size particle studied (< 53 μm in diameter)

2) The concentration of Pu and Am in the soil samples does not appear to be homogeneous; consequently, the results presented in this report (determined by assuming a uniform extent of contamination in the soil samples and determining the amount of Pu and Am left in the soil samples after leaching) are preliminary. Analyses of both phases (liquid and solid) in the leaching experiments is underway. These results will be presented in a future report.

3) Several promising leaching schemes can be identified from inspection of the results presented. The most efficient extractant was sodium citrate with most of the oxidizing or reducing agents tested. The overall best extraction of Pu and Am from the RFP soil was obtained with sodium citrate and $Na_2S_2O_4$.

4) Citric acid was also effective as an extractant with most additives, but it appears to be more effective for Pu removal than for Am and no better than sodium citrate for Pu.

5) Other complexants that show some promise are EDTA, especially with oxidizing agents, DTPA, and nitrilotriacetic acid.

6) As can be concluded by inspection of the results presented some of the leaching schemes utilized can extract 50% or more of the Am and Pu in the contaminated soils (without any optimization) which is very encouraging.

7) Comparison of the removal of Pu from RFP soils with the removal of Pu from soils from the NTS (utilizing similar leaching schemes) shows agreement.

8) The results presented here have positive implications for potential *in situ* treatment techniques (such as chemically-enhanced steam stripping). These results indicate that it is possible to leach Pu and Am from contaminated soils (at room temperature). It is expected that the extracting efficiency of the leaching schemes presented would increase at higher temperatures (such as the ones utilized during steam stripping). Consequently, future experiments should test this hypothesis by conducting leaching experiments at high temperatures using batch and column experiments.

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